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EFFECT OF PRESSURE AND TEMPERATURE ON PROPERTIES OF HOT-PRESSED GEOPOLYMER FOR IMMOBILIZATION OF RADIOACTIVELY CONTAMINATED WOOD ASH

Y.O. Svitlychnyi[1,2], S.Yu. Sayenko[1], B.G. Shabalin[2], V.A. Shkuropatenko[1], H.O. Kholomieiev[1], O.V. Pylypenko[1]

[1]National Science Center "Kharkiv Institute of Physics and Technology", Kharkiv, Ukraine
E-mail: esvitlychniy@i.ua;

[2]State Institution "The Institute of Environmental Geochemistry of National Academy of Sciences of Ukraine", Kyiv, Ukraine
E-mail: b_shabalin@ukr.net

The geopolymer based on an alkali-activated mixture of fly ash, blast furnace slag and metakaolin was produced and tested as a matrix for immobilizing the wood ash, obtained by pyrolysis of inactive pine, simulating radioactively contaminated wood from the Chernobyl zone. Ukrainian raw materials, hot pressing and casting were used to produce samples. The rheological properties of pastes, mechanical characteristics and structures of hardened samples were analyzed. Optimized hot pressing parameters: T = 80 °C and P = 60 MPa facilitate to form dense geopolymers containing 10…40 % wood ash with increased compared to casted samples properties, which meet the European Waste Acceptance Criteria.

INTRODUCTION

Intensive researches are focusing now in the world on the development of suitable materials for radioactive waste (RAW) immobilization. Aluminosilicate geopolymers (GP) combine acceptable physical and mechanical properties, possess good chemical resistance to different aggressive media and occupy one of the leading places among studied binders because they are considering as the most promising materials to replace ordinary Portland cement (OPC) [1–5].

The advantage of GP technology over OPC production is the absence of high-temperature firing and carbon dioxide emissions during the decomposition of carbonate raw materials, as well as the ability to utilize waste from various industrial sectors: fly ash, blast furnace slag, etc. [6–8].

GP are widely used as construction, thermal insulation and roofing materials, as well as in the manufacture fire-resistant composites and catalysts. Furthermore, GP are regarded as promising materials for the RAW immobilization and the use of such materials will significantly improve environmental safety and the reliability of waste long-term storage in future.

NSC KIPT systematically conducts research on the development of radiation-resistant ceramic, glass-ceramic and binder materials and significant attention is devoted to identifying promising formulations and finding new solutions for RAW immobilization [9–13]. Our experiments with GP testing were started in the framework of the Horizon-2020 (PREDIS project), which aimed the development and implementation of activities for pre-disposal treatment of RAW streams other than nuclear fuel and high-level radioactive waste. The main objectives of the research on the direct conditioning process were to select the most promising formulations for further study and development, to evaluate the stability of the selected formulations and to consolidate the reference formulations through testing using real waste, as well as to investigate the possibility of scaling up the results of previous pilot tests [14].

As a result of selection activity, the method for incorporating liquid RAW into GP was developed and optimized by NSC KIPT in terms of compliance with European Waste Acceptance Criteria (WAC). The obtained GP based on fly ash (FA), blast-furnace slag (BFS) and metakaolin (MK) mix is characterized with accepted workability, setting time – 20 h, dense, homogeneous structure and compressive strength – 32 MPa [15].

GP fabricated according developed formulation was tested as matrix for surrogated solutions (oils, solvents), which were used to simulate the liquid organic RAW. As a result of the direct conditioning process, samples containing up to 30% by volume of various oils are characterized with a dense, homogeneous structure and compared to samples without oils, have acceptable properties that are not significantly lower after 28 days of curing.

As a result of the Chernobyl accident, there was a large-scale regional release of radionuclides into the atmosphere, which caused radioactive contamination of the environment. Thanks to financial support from the EC, special equipment (an incinerator) was built in Chernobyl zone to burn radioactively contaminated wood. Wood ash is temporarily stored in special sealed containers, but it cannot be stored in this form for a long time. Therefore, the challenge of immobilizing the radioactive ash into a solid, stable matrix suitable for long-term storage is extremely relevant.

It should be noted that a number of scientific works and reviews on innovative GP have recently been published. One of the new routes to improve the GP characteristics is realizing with application of the hot

pressing (HP) method, where the pressure (P) and temperature (T) of process play an important role [16–20].

The aim of this work is to evaluate the effect of pressure and temperature on the immobilization of incinerated radioactive wood ash in GP matrix.

EXPERIMENTAL METHODOLOGY

In the research Ukrainian raw materials were used. MK was obtained by calcination of kaolin (Zaporozhye region) at 800 ºC. FA (class F) and ground granulated BFS were supplied by thermal power plant Burshtyn (Ivano Frankivsk region) and CEMBUDSERVICE Co (Kam'yanske, Dnipro region) respectively.

Potassium hydroxides and potassium liquid glass were used as alkaline activators. Preliminary tests were conducted with GP without RAW simulants to optimize the parameters and after the research was performed with addition of simulant in GP matrix.

Inactive incinerated wood ash was used as the simulant of RAW (radiation-contaminated wood from Chernobyl zone). Firstly, the dry mixture of raw materials: FA – 34 wt.%, BFS – 20 wt.%, and MK – 14 wt.%, was prepared and secondly, the alkaline activator based on liquid mixture of components: distilled water – 12 wt.%, potassium hydroxide – 9 wt.% and potassium glass – 11 wt.% was prepared. Then the dry mixture was added to activator and mixed for 20 min to make a fresh GP paste, which was placed into mold and installed to lab hot press. Then HP was realizing with T = 60…150 °C and P = 50…70 MPa. Formed GP were pressed out and cured at room temperature for 28 d.

The first step of the research was to establish the optimal parameters of HP: pressure and temperature for GP producing without wood ash. Based on the results obtained from optimizing the parameters for GP producing, the compositions of the test samples were chosen to investigate the possibility of immobilizing wood ash into the GP matrix. For the corresponding experiments, wood ash was added to the GP paste in amounts ranging from 10 to 40 wt.%.

The scheme of HP treatment GP paste and lab scale HP facility are presented in Figs. 1 and 2, respectively.

A series of tests were performed to evaluate the fresh pastes and hardened materials properties. Pastes and hardened samples were tested regarding bleeding, setting time, workability, determined by viscosity measurement, and compressive strength in terms of compliance with European WAC.

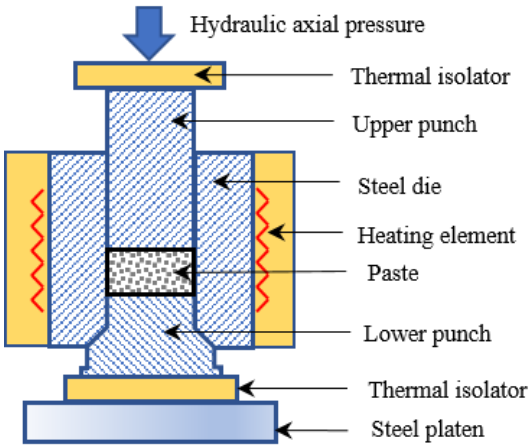


Fig. 1. Scheme of HP treatment of geopolymer paste

The viscosity of the paste was determined using a Brookfield LVDV-II+ Pro rotational viscometer. Compressive strength was determined in accordance with DIN EN 196-1 using a testing press. Some characteristics were evaluated visually.



Fig. 2. Lab scale HP facility

Studies of the macrostructure of the materials were performed using an optical microscope MBS-10 and studies of the microstructure of the GP samples were conducted using a scanning electron microscope Superprobe 733 (JEOL, Japan).

RESULTS AND DISCUSSION

The effect of pressure and temperature on compressive strength of GP samples is illustrated in the Fig. 3.

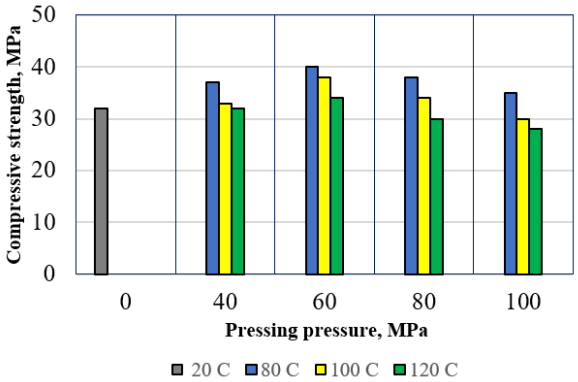
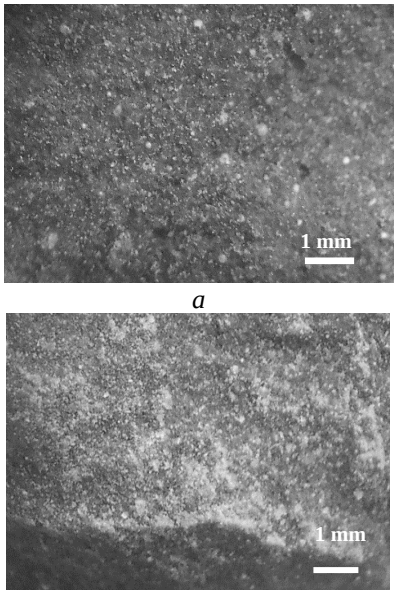


Fig. 3. Effect of pressure and temperature on compressive strength of samples

Samples produced using the HP method are characterized with higher compressive strength compared to samples produced by casting (20 °C). When the pressing pressure is increased from 40 to 60 MPa, the strength of the samples increases at all applied temperatures (80…120 °C) and reaches a maximum strength of 40 MPa at 80 °C. With a further increase in pressing pressure to 80 and 100 MPa, the strength of the samples decreases. The probable cause of the decrease in strength at temperatures above 80 °C is the increased evaporation of the alkaline activator solution, which prevents the formation of the advanced geopolymer.

The macrostructure of samples produced by HP method (P = 60 MPa, T = 80 °C) compared to casted samples (T = 20 °C) is shown in the Fig. 4.

The structure of the samples obtained using HP method is similar to the structure of casted samples, mostly dense and homogeneous. The only difference is that casted samples contain more pores and other defects with size ≤ 0.5 mm.



a

b

Fig. 4. The macrostructure of samples produced by HP method (a) compared to casted samples (b)

Thus, the influence of temperature and pressing pressure on the properties of GP was determined, and the optimal parameters for obtaining samples with advanced strength were established: T = 80 °C, P = 60 MPa.

It is worth noting that the results of our study and the established parameters for the production of improved GP are consistent with the findings of a number of researchers [21-24], which confirms the validity of our conclusions.

Based on the obtained results with optimized HP parameters for GP producing, the compositions of the test samples were chosen to investigate the possibility of immobilizing wood ash (10…40 wt.%) into the GP matrix. In accordance with the WAC for immobilizing matrix, the viscosity of the paste with wood ash added was investigated.

Rheograms of the optimal GP paste and the paste with varying wood ash content are presented in Fig. 5.

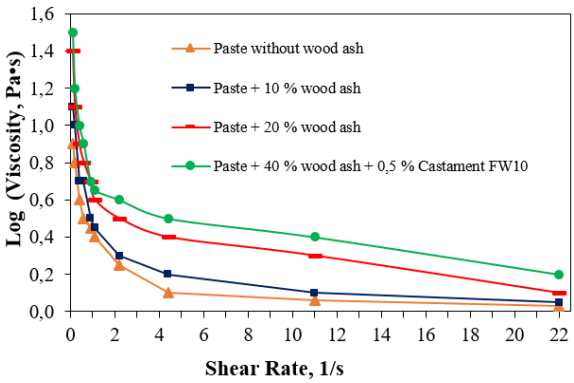


Fig. 5. Rheograms of the optimal GP paste (without wood ash) and the paste with varying wood ash content

It was determined that pastes of all compositions are characterizing by decreasing viscosity at increasing shear rate, which indicates the pseudoplastic behavior of the tested materials. Samples of GP paste without wood ash as well as with 10 and 20% of wood ash content are identified by similar behavior. As the wood ash content increases, the viscosity of the paste increases correspondingly at all shear rates studied.

The paste with a 40% of wood ash is described with unsatisfactory flowability, and it was not possible to determine its viscosity using the viscometer. Therefore, surfactant Castament FW-10 (BASF) based a polyethylene glycol was used to improve flowability. The addition of the Castament FW-10 into the paste results in a significant decrease of viscosity (see Fig. 5) and allows to obtain higher quality samples. The deflocculating action of Castament FW-10 is the typical effect of surfactants that causes a decrease in surface or interfacial tensions.

Based on the results of the studies conducted, GP samples were obtained with additions of 10, 20, and 40 wt.% wood ash, using both the ordinary casting method and HP treatment, with the following optimized parameters: T = 80 °C, P = 60 MPa.

Photo of casted samples (cubic) and treated with HP method (cylindric) for analytical researches are illustrated in Fig. 6.

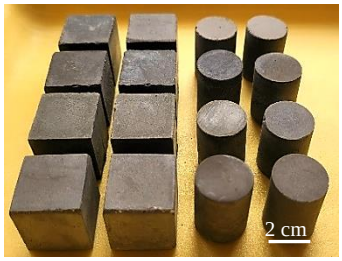


Fig. 6. Photo of casted samples (cubic) and treated with HP method (cylindric), from bottom to top: without wood ash (1 row), with 10% of wood ash (2 row), with 20% of wood ash (3 row), with 40% of wood ash (4 row)

The mechanical properties of samples produced by casting and HP method for comparison are reported in Table.

Mechanical properties of samples produced by casting and HP method

Method of production	Wood ash content, wt.%	Density, g/cm^3	Compressive strength, MPa	
			7 d	28 d
Casting	0	1.96	23	32
	10	1.85	18	28
	20	1.76	14	22
	40	1.64	10	16
HP	0	2.05	30	40
	10	1.92	22	32
	20	1.84	17	26
	40	1.73	12	20

GP samples of all compositions, which were produced using HP treatment and tested after 7 and 28 days, are presented with higher density and strength values compared to casted samples. With an increase in

wood ash content from 10 to 40%, a significant reduction in properties of all compositions is observed.

Regarding the compressive strength data, all samples without and with wood ash added (10…40%), casted and treated by HP measured > 5 MPa at 28 days, which complies with WAC and confirms the robustness of the formulation.

SEM observations were conducted on GP samples with 40% of wood ash, produced by HP treatment, compared to casting method, as reported in Fig. 7. Both GP samples are characterized with identical microstructure: very dense and homogeneous.

Fig. 8 demonstrates the SEM image and results of EDS of the sample with 40% wood ash, produced by HP.

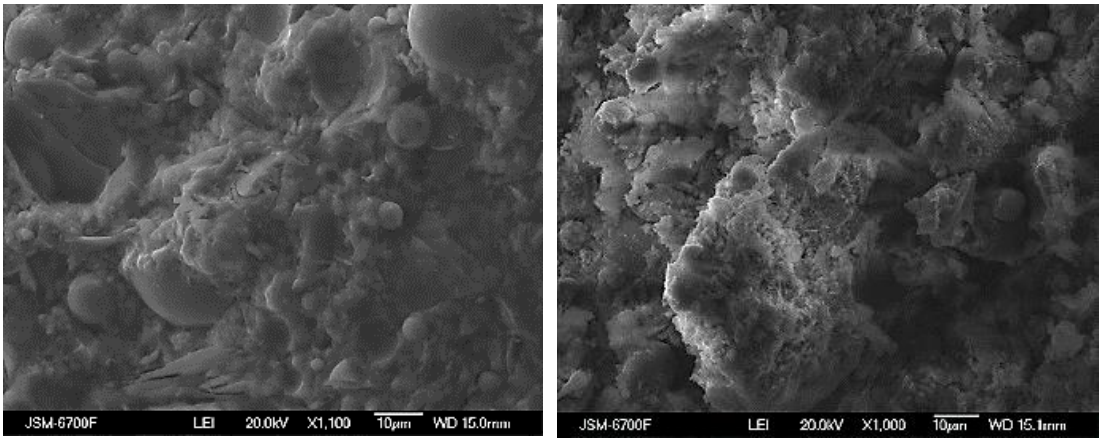
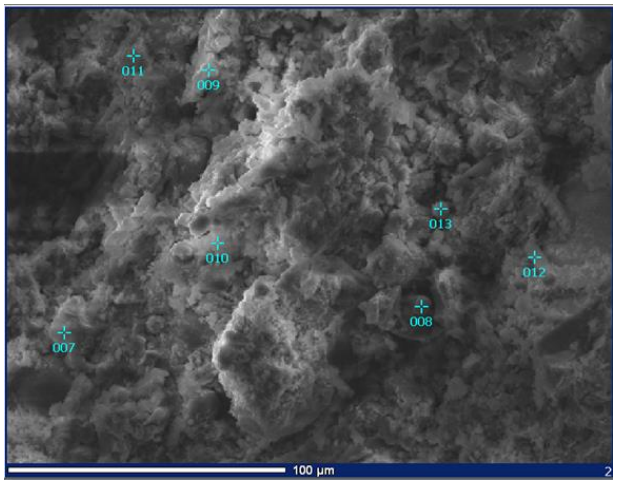


Fig. 7. SEM of GP samples with 40% of wood ash, produced by HP treatment (right), compared to casting method (left)



Memo	Na2O	MgO	Al2O3	SiO2	SO3	K2O	CaO	TiO2	Cr2O3	MnO	FeO	CoO	NiO	CuO	ZnO
007	0.31	3.44	7.82	59.61	1.54	17.19	7.29	0.20	0.10	0.11	1.77	0.10	0.26	0.27	0.00
008	0.29	1.96	3.01	68.03	0.35	18.87	5.42	0.16	0.23	0.13	0.87	0.21	0.00	0.16	0.30
009	0.28	2.53	20.66	51.83	0.71	7.12	13.45	0.20	0.14	0.42	2.01	0.26	0.39	0.00	0.00
010	0.00	2.06	16.21	44.20	1.15	10.42	22.59	1.05	0.66	0.34	1.24	0.02	0.02	0.05	0.00
011	0.59	8.35	1.83	47.87	1.41	15.55	23.35	0.00	0.00	0.51	0.49	0.00	0.00	0.00	0.04
012	0.59	1.66	5.36	83.72	0.31	3.66	3.74	0.30	0.00	0.04	0.34	0.06	0.09	0.12	0.00
013	1.20	2.32	32.24	54.71	0.00	3.60	1.64	0.28	0.00	0.02	3.48	0.04	0.03	0.18	0.26

Fig. 8. SEM and EDS of the sample with 40% wood ash added produced by HP

Elemental analysis of microstructural sections of the sample have been displayed the presence of spherical fly ash particles with a low calcium content (areas 7, 8) and wood ash particles with a high calcium content (areas 10, 11); the high calcium content in wood ash was previously observed and reported in the work [12]. As can be seen, the wood ash particles are quite firmly bound to the GP matrix, which is demonstrated in areas 9 and 13.

CONCLUSIONS

Experimental studies were conducted on the development of GP composite based on Ukrainian raw materials: fly ash, blast furnace slag, metakaolin, as well as liquid potassium glass and potassium hydroxide, using HP method and ordinary casting for application in the immobilization technology of incinerated radioactive wood ash from Chernobyl zone.

According to the results of the rheological test, it was determined that both GP pastes without and with wood ash added are characterizing by decreasing viscosity at

increasing shear rate, which indicates the pseudoplastic behavior of the tested materials. As the wood ash content increases, the viscosity of the paste increases correspondingly at all shear rates studied. The effective impact of the addition of a surfactant Castament FW-10, to improve the fluidity and the homogeneity of the paste with 40 wt.% of wood ash added was demonstrated.

Based on the analysis of the influence study of temperature and pressure on the GP characteristics the optimal parameters of HP method were established: T = 80 °C and P = 60 MPa, which initiate the highest compressive strength – 40 MPa. It was shown that the formed by HP method GP samples with 10…40% wood ash content, are characterized by a dense and homogeneous structure and acceptable property indices, which exceed those of samples produced by casting.

The results of this research have been demonstrated the advantages of using the HP method over ordinary casting, as well as its potential for future application in the immobilization technology of incinerated radioactive wood ash.

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ВПЛИВ ТИСКУ ТА ТЕМПЕРАТУРИ НА ВЛАСТИВОСТІ ГЕОПОЛІМЕРУ, ЩО ОТРИМАНИЙ МЕТОДОМ ГАРЯЧОГО ПРЕСУВАННЯ, ДЛЯ ІММОБІЛІЗАЦІЇ РАДІОАКТИВНО ЗАБРУДНЕНОЇ ЗОЛИ ДЕРЕВИНИ

Є.О. Світличний, С.Ю. Саєнко, Б.Г. Шабалін, В.А. Шкуропатенко, Г.О. Холомєєв, О.В. Пилипенко

Геополімер на основі лужно-активованої суміші золи виносу, доменного шлаку та метакаоліну було виготовлено та випробувано в якості матриці для іммобілізації золи деревини, яку отримано шляхом піролізу неактивної сосни, що імітувала радіоактивно забруднену деревину з Чорнобильської зони. Для виготовлення зразків використовували українську сировину, гаряче пресування та лиття. Проведено аналіз реологічних властивостей паст, механічних характеристик та структур затверділих зразків. Оптимізовані параметри гарячого пресування: T = 80 °C та P = 60 МПа, сприяють утворенню щільних геополімерів, що містять 10…40% золи деревини, з покращеними, як порівняти з литими зразками властивостями, які відповідають Європейським критеріям приймання відходів.